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INTRODUCTION

TO THE

STUDY OF PHYSICAL
CHEMISTRY

BY

SIR WILLIAM RAMSAY, K.C.B., F.R.S.

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INTRODUCTION TO THE STUDY
OF PHYSICAL CHEMISTRY

TEXT-BOOKS OF PHYSICAL CHEMISTRY.

EDITED BY SIR WILLIAM RAMSAY, K.C.B., F.R.S.

THE progress of Physical Chemistry is now so rapid, its domain is so extensive, and the number of journals devoted to its exposition is so great, that it has appeared desirable to issue a series of volumes, each of moderate compass, and each dealing with one branch of the subject. The rate of advance in various branches of the subject is not equal; while, for example, the basis of the science remains comparatively stationary (for methods of determining atomic and molecular weights, and the classification of compounds undergoes little modification), rapid progress is being made in other branches. Hence it has been thought proper to issue several short manuals, so that each individual one may be frequently brought up to date, independently of others. In this way, a statement of what is known on each subject will be made accessible to students and investigators. The subject has been divided as follows, among the authors mentioned:—

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The order in which these parts will be issued is not that given; for some subjects are more easily treated of than others; but it is hoped that all the volumes will be ready for the press before December, 1904.

¹ This is the General Introduction to the series, and it also appears in Mr. Findlay's book on the Phase Rule.

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To define exactly the provinces of the Sciences of Physics and Chemistry is not easy. The definition that the object of Chemistry is the study of the changes which matter undergoes during the formation and decomposition of compounds, while that of Physics has reference to changes which affect matter independently of its composition, hardly meets the case. It is true that it is possible to deduce certain laws relating to the properties of matter, which are valid irrespective of the chemical composition of the matter; but such laws deal with the behaviour of matter in motion, or acted on by gravitational or other forces which apply only to matter in bulk. For example, Newton's law of "gravitation"—that two masses approach each other as if impelled by a force which varies directly as their masses, and inversely as the square of the distance which separates them—takes no cognizance of the nature of the matter which attracts: the law is not affected by the chemical composition of the acting bodies. Similarly, the laws dealing with inertia are true, whatever the nature of the matter to which these laws apply. But when phenomena relating to change of volume by pressure, to the surface phenomena of liquids, to electricity, to heat, light, and other forms of wave-motion, are considered, the laws deduced from them contain specific

constants, so that their numerical application can be made only when these specific constants are known. Thus although the same differential equation governs the flow of heat in all kinds of matter, it contains a specific constant, viz. the "conductivity;" and as this varies for different kinds of matter, the chemical nature of the substance becomes a factor in the problem.

The birth of physical chemistry may be said to date from the recognition of this fundamental idea; where the laws or generalizations regarding properties of matter depend not merely on the masses or rates of motion of the objects considered, but also on their composition and chemical nature, their consideration falls under the heading "Physical Chemistry."

It was not until the middle of the nineteenth century that this began to be recognized; although many facts had been discovered, and many laws deduced before that date, physical chemistry was not differentiated from physics, on the one hand, and chemistry on the other, until the dependence of certain physical phenomena on the chemical composition of the objects under consideration had become obvious.

The attempts of the ancients to interpret the facts around them led to little of value. For long mankind was contented to observe certain phenomena, and to utilize them for industrial purposes, if they were found suitable. Yet in all ages "philosophers," or lovers of wisdom, as they liked to call themselves, have endeavoured to "explain" certain observed facts. This word "explain" is frequently used without any very definite view of its signification. It may be defined as "to state the unknown in terms of the known," and this process was facilitated by grouping observed facts into similars and dissimilars. The "properties of matter," for example, were "explained" by the Greeks and their predecessors by the theory that all matter partook of the nature of fire, air, earth, and water—the so-called "elements"—in greater or less proportion. But this

hypothesis, which, so long as only the solidity, fluidity, or gaseous nature of matter was in question, appeared reasonable, failed to throw light on the changes which matter undergoes during combustion. And in the seventeenth century, Boyle, in his *Sceptical Chymist*, pointed out that it was impossible to explain the existence of the numerous chemical substances known in his day, or the transformations which they may be made to undergo, by this ancient hypothesis. The word "element" was otherwise defined by Boyle as the constituent of a compound body; and he denied that the properties of matter could be modified by assimilating the qualities of fire, air, earth, and water.

In the light of this revolution of thought it became of importance to determine which of the numerous forms of matter were to be regarded as elementary and which as compound, or composed of two or more elements in a state of combination; and also to produce such compounds by causing the appropriate elements to combine with each other.

The progress of science was, however, retarded by the existence of certain preconceived notions, which had first to be disproved before advance was possible. The first of these was the failure to recognize the material nature of air, and to differentiate between one gas and another. Although Boyle himself discovered the most important law, which goes by his name, and which deals with the compressibility of air, and although the fact that air possesses weight had been foreshadowed by Jean Rey, physician to the court of France, and by Torricelli, the inventor of the barometer, it was not until experiments were made by Black, in 1752, on carbonic acid that it was distinctly recognized that gases may differ in kind, and that each possesses its own particular density. The investigations of Scheelè, Priestley, and Cavendish in the succeeding years showed that many kinds of gas exist; and in the following century Faraday, by liquefying ammonia, chlorine, and other

gases, proved that a gas is merely a vapour at a high temperature; and Andrews, by his investigation on the critical temperature of carbon dioxide, showed that above that temperature that gas cannot be liquefied by compression alone. Towards the end of the nineteenth century, Pictet and Cailletet succeeded in liquefying oxygen; Wroblewski, Olszewski, Kammerlingh Onnes, and Dewar prepared liquid oxygen and air on a fairly large scale; and the more recently invented machines of Hampson and Linde have much facilitated the process of liquefaction. Olszewski was the first to liquefy hydrogen in minute quantity; and on a larger scale, Dewar and, later, Travers have succeeded in preparing liquid hydrogen in bulk. The only gas which has as yet resisted liquefaction is helium.

The second fallacy was an inverted notion of the phenomena of combustion. It had long been held, probably since the eighth century, that the property of burning which many substances possess was due to the sulphur which it was supposed that they contained. At that date, what were later called "hypostatical (or underlying) principles" were added to the four elements, as capable of modifying the nature of matter. These were: salt, which conferred fixity in the fire; mercury, which conferred volatility and metallic lustre; and sulphur, which carried with it combustibility. This idea was somewhat modified by Becher, who conceived all combustible matter to contain a *terra pinguis*, or fatty earth. His pupil, Stahl, having observed that it was possible to confer combustibility on many substances which had lost that power on having been burned, by calcining them in absence of air with bodies such as charcoal, wood, coal, flour, etc., which themselves possessed the property of burning, extended, about the year 1690, the use of the idea of "sulphur," or *terra pinguis*, and at the same time changed its name to "phlogiston." We see here the conception of the substantial nature of flame, which will be alluded to later on. This phlogiston was not merely a

principle which was lost during combustion ; it was at the same time possible to transfer it from a body rich in it to one containing little or none. From this point of view, metals were regarded as substances rich in phlogiston ; when they were calcined in the air, the remaining "calces" were looked upon as metals from which phlogiston had escaped, but by heating such "calces" with charcoal or other substances rich in phlogiston, that principle was transferred to the "calces," and metals were reproduced. The metals were therefore considered to be compounds of their calces with phlogiston. Other substances, such as phosphorus or sulphur, when burned, gave acid liquids, to which it was not so easy to add phlogiston ; but even they could be phlogisticated ; and sulphur and phosphorus were regarded as compounds of sulphuric and phosphoric acids with phlogiston.

The discovery of oxygen in 1774 by Priestley and by Scheele, and the explanation of its functions by Lavoisier during the ten following years, revolutionized the method of regarding combustion. It was then recognized that combustion is union with oxygen ; that an earth, or "calx," is a compound of a metal with oxygen ; that when a metal becomes tarnished and converted into an earthy powder, it is being oxidized ; that the resulting oxide, when heated to redness out of contact with air with charcoal or carbon, or with compounds such as coal, flour, and wood, of which carbon is a constituent, gives up its oxygen to the carbon, forming an oxide of carbon, carbonic oxide, on the one hand, or carbonic "acid" on the other, while the metal is reproduced in its "reguline" or metallic condition ; and that the true elements are metals, carbon, phosphorus, sulphur, and similar bodies, and not the products of their oxidation.

The third mistaken notion was the material nature of heat. The fact that flames were actually seen issuing from burning bodies early led to the view that they were material objects ;

and fire was therefore regarded as one of the "elements." Even after the overthrow of the ancient notions of combustion, it was believed that, although devoid of weight, heat was a substance. The earlier treatises often began with "the three imponderables"—heat, light, and electricity. Under the name of "caloric" this view was long held regarding heat. Towards the end of the eighteenth century Count Rumford had noticed, at Munich, that an apparently inexhaustible supply of heat could be generated by the boring of cannon, and that if bored under water the rise of temperature was such that the water could be kept in a state of ebullition. He argued, therefore, that the heat was not previously in combination with the iron of the cannon, or of the drill, but that it was produced by friction. At a later date Davy succeeded in melting two pieces of ice by rubbing them together.

That bodies possess a certain capacity for, or power of holding, heat was discovered by Leslie and Black, who investigated the specific heats of many different substances. Black also made experiments on the quantity of heat absorbed by different substances when they melt or evaporate, and introduced the conception of "latent" heat, *i.e.* heat which is possessed by a body, but which is insensible to a thermometer. The first table of capacities for heat was published by Kirwan in 1780: though Crawford had published the result of attempts to determine the specific heats of gases in 1779.

In 1801 Dalton published in the memoirs of the *Manchester Literary and Philosophical Society*, 5, p. 515, an account of experiments which proved that while the compression of gases was attended by an evolution of heat, their expansion rendered them colder; and Leslie attempted to introduce quantitative measurement. In 1807 Gay-Lussac applied the temperature changes resulting from the compression of gases to the measurement of their specific heats; and in 1816 Laplace had differentiated between the specific heat of a gas

at constant volume and under constant pressure, and had applied his results to Newton's equation relating to the velocity of sound. The explanation was first given, however, by Julius Robert Mayer. While former investigators held the idea that the phenomena were to be explained by the view that on compressing a gas caloric escaped, as water does when a sponge is squeezed, and on allowing it to expand the heat enters and again fills the pores of the gaseous matter, Mayer regarded the heat produced as the *equivalent of the work expended on the gas*. From the known alteration of temperature which a gas undergoes on expanding and the resulting heat which it absorbs on the one hand, and from the work which the gas can do during expansion on the other, he attempted to calculate the equivalence of heat and work. Almost at the same date J. P. Joule made quantitative experiments, in which he measured the rise of temperature of a known weight of water when it was set in violent agitation by a stirring apparatus driven by a descending weight. The doctrine of the existence of heat as a subtle form of matter thus gradually became extinct; and heat is now regarded when it is associated with matter as a state of motion of the particles of matter, atomic or molecular; and when "radiant"—*i.e.* during the condition of transit from one body to another (in which state the name "heat" is perhaps inapplicable)—as a state of strain in an imaginary medium, the ether, the strain being passed from place to place in the form of waves.

It was not until these misconceptions—the immaterial nature of gases, the inverted notion of combustion, and the material nature of heat—had been removed, that progress in physical chemistry became possible. But even with their removal the way was not clear, for it was necessary that a working hypothesis regarding the nature of matter should first have been formed. This was primarily due to John Dalton, a Manchester schoolmaster.

It had been noticed by Wenzel, by Richter, by Wollaston, and by Cavendish towards the close of the eighteenth century that the same compounds contain the same constituents in the same proportions, or, as it is expressed, "possess constant composition." Wollaston, indeed, had added to this the further fact that when the vegetable acid, oxalic acid, is combined with potash it forms two compounds, in one of which the acid is contained in twice as great amount relatively to the potash as in the other. The names monoxalate and binoxalate of potash were applied to these compounds to indicate the respective proportions of the ingredients. Dalton conceived the happy idea that by applying the ancient Greek conception of atoms to such facts, the relative weights of the atoms could be determined. Illustrating his views with the two compounds of carbon and hydrogen, marsh gas, and olefiant gas, and with the two oxides of carbon, carbonic oxide and carbonic "acid," he regarded the former as a compound of one atom of carbon and two of hydrogen, and the latter as a compound of one atom of carbon and one of hydrogen, and similarly for the two oxides of carbon. Knowing the relative weights in which these elements enter into combination, he deduced what he supposed were the relative weights of the atoms.

Dalton's work was first expounded by Thomas Thomson, Professor at Glasgow, in his *System of Chemistry*, published in 1805, and subsequently in Dalton's own *System of Chemical Philosophy*, the three volumes of which were published in 1808, in 1810, and in 1827.

The determination of these "constants of Nature," the atomic weights, was at once followed out by many chemists, Thomson among the first. Chief among the chemists who pursued this branch of work was Jacob Berzelius, a Swede, who devoted his long life (1779-1848) to the preparation of compounds, and to the determination of their composition, or, as it is still termed, the determination of the "atomic weights"

—more correctly *equivalents*—of the elements of which they are composed. It is to him that we owe most of our analytical methods, for prior to his time there were few, if any, accurate analyses. Although Lavoisier had devised a method for the analysis of compounds of carbon, viz. by burning the organic compounds in an atmosphere of oxygen contained in a bell-jar over mercury, and measuring the volume of carbon dioxide produced, as well as that of the residual oxygen, Berzelius achieved the same result more exactly and more expeditiously by heating the substance, mixed with potassium chlorate and sodium chloride, and thus estimating the hydrogen as well as the carbon. This process was perfected by Liebig. Berzelius, however, was able to show that compounds of carbon, like those of other elements, are instances of combination in constant and multiple proportions.

In 1815 two papers were published in the *Annals of Philosophy* by Dr. Prout, which have had much influence on the progress of chemistry. They dealt with the figures which were being obtained by Thomson, Berzelius, and others, at that time supposed to represent the “atomic weights” of the elements. Prout’s hypothesis, based on only a few numbers, was that the atomic weights were multiples of that of hydrogen, taken as unity. There was much discussion regarding this assertion at the time, but as it was contradicted by Berzelius’s numbers, the balance of opinion was against it. But about the year 1840 Dumas discovered an error in the number (12·12) given by Berzelius as the atomic weight of carbon; and with his collaborator, Stas, he undertook the redetermination of the atomic weights of the commoner elements—for example, carbon, oxygen, chlorine, and calcium. This line of research was subsequently pursued alone by Stas, whose name will always be remembered for the precision and accuracy of his experiments. At first Stas inclined to the view that Prout’s hypothesis was a just one; but it was completely disproved by

his own subsequent work, as well as by that of numerous other observers. It is, nevertheless, curious that a much larger proportion of the atomic weights approximate to whole numbers than would be foretold by the doctrine of chances; and perhaps the last has not been heard of Prout's hypothesis, although in its original crude form it is no longer worthy of credence.

To Dalton the smallest portions of matter, whether consisting of single atoms, as that of oxygen, O, or of compounds, such as water, to which he ascribed the composition HO, were alike regarded as atoms. This view, however, conflicted with experimental data arrived at by Gay-Lussac in the year 1808. In conjunction with Humboldt, Gay-Lussac had rediscovered about three years before what had previously been established by Cavendish, namely, that, as nearly as possible, two volumes of hydrogen combine with one volume of oxygen to form water, the gases having been measured at the same temperature and pressure. Humboldt suggested to Gay-Lussac that it would be well to investigate whether similar simple relations obtain between the volumes of other gaseous substances when they combine with each other. This turned out to be the case; it appeared that almost exactly two volumes of carbonic oxide combine with one volume of oxygen to form two volumes of carbonic acid gas; that equal volumes of hydrogen and chlorine unite to form hydrochloric acid gas; that two volumes of ammonia consist of one volume of nitrogen in union with three volumes of hydrogen, and so on. From such facts Gay-Lussac was led to make the statement that the weights of equal volumes of gases, whether simple or compound, and therefore their densities, are proportional to their empirically found combining or atomic weights, or to rational multiples of the latter. Gay-Lussac regarded this discovery of his to be a support for the atomic theory, but it did not accord with many of the then received atomic weights.

The assumption that equal volumes of gases contain equal numbers of particles, or, as they were termed by him, *molécules intégrantes*, was made in 1811 by Avogadro, Professor of Physics at Turin. This theory, which has proved of the utmost importance to the sciences of both chemistry and physics, had no doubt occurred to Gay-Lussac, but had been rejected by him for the following reasons: A certain volume of hydrogen, say 1 cubic centimeter, may be supposed to contain the same number of particles (atoms) as an equal volume of chlorine. Now, these two gases unite in equal volumes. The deduction appears so far quite legitimate that 1 atom of hydrogen has combined with 1 atom of chlorine. But the resulting gas occupies 2 cubic centimeters, and must therefore contain the same number of particles of hydrogen chloride, the compound of the two elements, as 1 cubic centimeter originally contained of hydrogen or of chlorine. Thus we have 2 cubic centimeters containing, of uncombined gases, twice as many particles as is contained in that volume after combination. Avogadro's hypothesis solved the difficulty. By premising two different orders of particles, now termed atoms and molecules, the solution was plain. According to him, each particle, or molecule, of hydrogen is a complex; it contains 2 atoms; the same is the case with chlorine. When these gases combine, or rather react, to form hydrogen chloride, the phenomenon is one of a change of partners; the molecule, the double atom, of hydrogen splits; the same is the case with the molecule of chlorine; and each liberated atom of hydrogen unites with one of chlorine, forming a compound, hydrogen chloride, which equally consists of a molecule, or double atom. Thus 2 cubic centimeters of hydrogen chloride consist of a definite number of molecules, equal in number to those contained in 1 cubic centimeter of hydrogen plus those contained in 1 cubic centimeter of chlorine. The case is precisely similar if other gaseous compounds be considered.

Berzelius was at first inclined to accept the theory, and indeed went so far as to change many of his atomic weights to make them fit it. But later he somewhat withdrew from his position, for it appeared to him hazardous to extend to liquids and solids a theory which could be held only of gases. Avogadro's suggestion accordingly rested in abeyance until 1858, when Cannizzaro, Professor of Chemistry in Rome, published an essay in which all the arguments in favour of the hypothesis were collected and stated in a masterly manner.

Although as early as 1811 a distinction had been drawn between the two classes of particles, atoms, or single particles of elements, and molecules, or congeries of atoms, which may either be of one kind, in which case the molecule is one of an element, or of different kinds, in which case the molecule is one of a compound, yet, as we have seen, the theory was not accepted by chemists and physicists until after 1858.

Inasmuch as physical chemistry rests on the molecular hypothesis of matter, researches before that date referred merely to the physical properties of matter, and just conclusions could not be drawn from them. It is necessary, in many very important researches, to pay attention to data considered with reference to equal numbers of molecules, instead of to equal masses of substance; and relations really simple remained undiscovered until the molecular hypothesis was generally adopted.

Yet many interesting observations were made before the middle of the eighteenth century, of great value in themselves, although their true bearing was not evident until the molecular hypothesis of matter had been established.

For example, Boyle's law (1662) relating to the compressibility of air was found to apply with more or less accuracy to the compressibility of other gases; and Gay-Lussac's or Dalton's law (1808) dealing with the expansion of gases with temperature, also holds approximately for all gases. The progress of

science, however, has always been characterized by the discovery of approximate relations ; the divergence from exactness has generally at first been attributed to the imperfection of the observations ; and with improvements in apparatus, and with introduction of extreme conditions, these " laws " have always been discovered to be only approximately applicable ; to render them more in accordance with fact, modifications have had to be introduced which lessen the deviations ; but all that can be said of any so-called " laws " is that at the best they represent the results of measurement as accurately as the methods of experiment allow.

It is seldom the case that the causes of divergence can be fully discovered. Yet in some instances it is possible to surmise that the cause is known, even although the phenomena are too complicated to be solved numerically. The moon pursues an approximately elliptical course round the earth, due to her own proper motion and to the attraction of the earth ; yet the path is influenced by the attraction of the sun, and in a lesser degree by that of the planets, and in a still smaller by that of the fixed stars. Owing to the number and complexity of these various attractions, it is impossible to do more than approximate numerically to the actual path of the moon's orbit ; yet as many of these attractions are almost inconceivably minute, their effect may be disregarded. In such an instance the law of attraction is a simple one ; but with molecules, instead of masses, the laws of attraction have still to be discovered.

Applying this conception to Boyle's and Gay-Lussac's laws, it was soon discovered that they held only with approximation ; and in 1829 Dulong and Arago, and later Pouillet, Regnault, and Natterer, investigated the variation of the product of pressure and volume, which, according to Boyle's law, should remain constant so long as temperature is stationary. The investigation from a practical point of view was continued by Amagat, Mathias, Ramsay and Young, and others ; and from

the theoretical side by van der Waals, Clausius, and many others.

It is clear that Boyle's law can be valid only under one of two suppositions—either that the molecules of a gas themselves occupy no space, or that the molecules themselves are equally compressible with the gas. The last supposition has not recommended itself; it has been generally assumed that molecules are analogous to solid spheres, or solid particles of some definite form; and that as a great increase of pressure causes only a small decrease in the volume of a solid, so a "solid" molecule must be regarded as practically incompressible. That the compressibility of a gas must necessarily be less than that calculable from Boyle's law was predicted by Daniel Bernoulli (1738); but he did not recognize that another influence may be at work, tending to reduce the distance between the molecules, namely, their mutual attraction. The two conceptions were first introduced by van der Waals. But the simple formula devised by him does not accurately represent facts, and we are still ignorant of even approximately exact laws relating to the compressibility of gases, and their expansion on rise of temperature.

But certain gases, on rise of temperature, did not show even the approximate regularity in expansion demanded by Gay-Lussac's law. Bineau, who had determined the density of many vapours at relatively high temperatures, discovered that ammonium chloride, cyanide, hydrosulphide and carbonate as well as phosphoric chloride, and chloride and bromide of phosphonium possessed a vapour density only half as great as that implied by their formulæ. The explanation was given by Cannizzaro, Kopp, and Kekulé. They pointed out that these substances decompose when heated into simpler constituents—ammonium chloride, for example, into ammonia and hydrogen chloride—and that, on cooling, the original substance is reproduced. To this decomposition Deville gave the name

“dissociation.” Würtz, in 1865, traced the gradual dissociation and re-formation of amylene hydrobromide, and other instances followed; from that date the justice of the theory of reversible decomposition by heat, or dissociation, of certain substances was established.

It was for long believed that molecules exercise a repulsive action on one another, and tend to part company on account of this repelling force. And it was known that if forces of any kind suffer displacement in the direction of their action without external work being done, heat is evolved. It would follow, therefore, that if a gas were to expand without doing work, its temperature should rise. Joule and Thomson (Lord Kelvin) showed, however, that on free expansion, most gases, instead of rising in temperature, undergo a small decrease. Hence there must be a small attractive, and not a large repulsive force between their particles.

Waterston, in 1845, attributed the pressure of a gas to the impacts of its molecules on the walls of the containing vessel, and on the manometer; while the temperature of the gas was conditioned by the rate of the motion of its particles. His work, however, remained unnoticed until attention was directed to it by Lord Rayleigh; and Krönig in 1856, and Clausius in 1857, following Joule in 1851, deduced from these hypotheses the “kinetic theory of gases,” by means of which, considering molecules as hard, smooth, elastic spheres, a mechanical explanation of Boyle’s and Gay-Lussac’s laws and of Avogadro’s hypothesis was given. By introducing numerical data for pressure and volume, the actual average velocity of a molecule of any gas could be calculated; and the measurements of Thomas Graham, published in 1833, of the rate of passage of gases through porous septa, and through narrow openings, were shown to approximate closely to numbers calculated on the basis of the relative average velocities of the molecules of different gases. It was also possible to calculate

the "free path," or average distance which a molecule traverses before collisions with other molecules; and also from measurements of the friction of gases in passing through capillary tubes, the relative diameters of the gaseous molecules, and consequently their volumes.

The attraction between the molecules of any gas decreases rapidly with rise of temperature; and with hydrogen gas, the boiling-point of which is about $20\cdot5^{\circ}$ absolute, or $-252\cdot5^{\circ}$ C., there is no sensible attraction at ordinary temperatures. Hence deviations from Boyle's law, in this case, by increase of pressure, are ascribable solely to the space occupied by the molecules relatively to that which they inhabit. It is therefore easy to calculate the actual volume of the "solid" molecules in any given volume of hydrogen gas; it appears to be somewhat less than $\frac{1}{10000}$ -th part of the space inhabited by hydrogen molecules at ordinary temperature and pressure. Knowing this, the actual diameter of a hydrogen molecule may be estimated as about $1\cdot6 \times 10^{-8}$ centimeter; or, otherwise expressed, there are present in 1 cubic centimeter of hydrogen 5×10^{19} molecules; or, again, the thousandth part of that volume, or 1 cubic millimeter of hydrogen gas contains about 50,000 million million molecules. The corresponding numbers for other gases can be calculated similarly from their relative diameters.

It has already been mentioned that Julius Mayer and Joule regarded heat as a form of energy (a word introduced by Rankine), and that Joule measured the equivalence between heat and work; and also that Clausius attributed the heat contained in gases, at least, to the state of motion of their molecules. The "First" and the "Second Law of Thermodynamics" stand in close connection with these conceptions. The first law is—that as heat and work are numerically interchangeable, and are both forms of energy, *energy is indestructible and uncreateable*. A loss of energy is only apparent; it is not really lost, but only appears in another form. The second is—that

heat cannot of its own accord pass from a colder to a warmer body; or in Thomson's (Lord Kelvin's) words: *It is impossible by means of any lifeless material contrivance to produce mechanical action from a given mass of matter by cooling it below the temperature of the coldest surrounding objects.* Still another method of expressing the same truth is—*A thermodynamic perpetual motion is impossible*; the perpetual motion being derived from a machine in which there is, it is true, neither creation nor destruction of energy, but a cyclical production of useful external work at the expense of a heat reservoir of constant temperature, or other source of "equilibrated" energy. These two laws have been productive of the greatest advances in physics and in chemistry; they may now be regarded as the fundamental bases of both sciences. The first law is involved in the determination of the ratio between the specific heat of gases at constant volume and at constant pressure. From this ratio a very important deduction has been drawn regarding the molecular complexity of gases, viz. that while certain gases, such as mercury-vapour and the gases of the argon group, consist of molecules of the simplest possible nature, that is, of single atoms, other gases, such as hydrogen, oxygen, nitrogen, and chlorine, consist of molecules composed each of two atoms. The conclusion as regards the last-mentioned gases had been arrived at from chemical considerations; but the proof was wanting, until corroborated by means of these physical aids. The subject has been fully worked out from the theoretical side by O. E. Meyer and by Boltzmann, whose work on the kinetic theory is also of great value.

Early researches of a physico-chemical nature were made in 1819 by Dulong, Director of the École Polytechnique at Paris, in conjunction with Petit, Professor of Physics there. These led to the discovery that equal amounts of heat are required to raise equally the temperature of solid and liquid elements, provided quantities are taken proportional to their atomic weight ;

in other words, equal numbers of atoms of elements have equal capacity for heat. By this means, an approximate estimate of the atomic weight of an element may be arrived at; and this, taken in conjunction with the exact determination of the equivalent of the element, may be made to yield a knowledge of the exact atomic weight. It would follow from this that as equal volumes of mercury gas and the gases of the argon group contain the same number of atoms (for the molecule is in their case identical with the atom), equal volumes should possess equal capacity for heat. If the molecular weight, or what is the same for these gases, the atomic weight, be expressed in grams, the specific heat at constant volume for such weights is 3; *i.e.* to raise the temperature of 4 grams of helium, 20 grams of neon, 40 grams of argon, 81.6 grams of krypton, 128 grams of xenon, or 200 grams of mercury-vapour through 1° , provided the gas is not allowed to expand, requires as much heat as could raise 3 grams of water from 18° to 19° C. And if the gas be allowed to expand, work will be done owing to the displacement of the atmosphere, so that other two heat units or calories are equivalent to that work; the specific heat "at constant pressure" is therefore 5, and the ratio between the two is as 3 : 5, or as $1 : 1\frac{2}{3}$. It is found that for gases with diatomic molecules, such as O_2 , N_2 , H_2 , etc., while the specific heat for equal atomic weights is 5 for constant volume, that for constant pressure is 7; the ratio between the two is as 5 : 7, or as $1 : 1.4$. Gases with more complicated molecules have higher specific heats; but the ratio between the atomic heat at constant volume and constant pressure is a smaller one, and does not display the same regularity as that between the atomic heats of monatomic and diatomic gases.

These relations are realizable by aid of the kinetic theory of gases, which postulates that the motion of the molecules is equivalent to their temperature. With monatomic molecules this motion is almost entirely translatory; with diatomic or

polyatomic molecules, on the other hand, there is not merely translatory motion from place to place in the containing vessel, but also there must be atomic motion within the molecule. The atomic motion is represented by the absorption of a certain amount of heat, when the temperature of the compound gas is raised. This view is due to Naumann (1876).

The specific heats of equal number of atoms of solids, on the other hand, if the atomic weights be expressed in grams, approximate to the number 6, nearly twice as great as that for monatomic gases at constant volume. It may be conjectured that the heat is partially accounted for by overcoming the restraint to which the molecules are subject in the solid state; and as the atomic heats are approximately equal, it may be argued that the molecules of all solid elements are subject to approximately equal restraint. As certain elements, namely, carbon, silicon, boron, and beryllium, are characterized by specially low atomic heats at ordinary temperature, it might be argued that their atoms are less restrained in their motion than those of the other solid elements. Yet, as Weber showed, at high temperatures their atomic heats become more normal; and it is difficult to see why a rise of temperature should increase any restraint due to natural attraction of atoms in the molecule. In compounds, too, as shown by Neumann (1831), Regnault (1840), Joule (1844), and Kopp (1864), the specific heats of equal numbers of molecules is approximately constant, though to some elements it would appear that exceptionally low atomic heats must be ascribed. Altogether the theory of the specific heats of solids and liquids is in an unsatisfactory state, and further investigations are much needed.

We have here an instance, however, of the necessity of taking into consideration the chemical constants of the substances investigated, *i.e.* atomic and molecular weights, in order to secure even approximate regularity. For the specific heats of elements and compounds present no regularity, unless they

be referred to atomic or molecular weights. Hence the determination of specific heats has been undertaken mainly by chemists; and the data obtained are among the earliest examples of physico-chemical constants.

The relations discovered by Gay-Lussac between the volumes of gases and their combining proportions stimulated others to investigate the question whether similar relations could not be found for liquids and solids. The first successful attempts were made by Hermann Kopp in 1842. The number of cubic centimeters of liquid at its boiling point, obtainable by condensing that volume of gas which contains the molecular weight of the liquid expressed in grams, was termed by Kopp the "specific volume." But it has since been found preferable to retain this term for the reciprocal of the density, *i.e.* for the volume occupied by one gram of the substance; and to substitute the term "molecular volume" for the constant suggested by Kopp. Working with carbon compounds, it was easy, by subtraction, to ascertain the difference in molecular volume for compounds differing by CH_2 , *i.e.* the difference between consecutive compounds in a homologous series, and so to arrive at values for individual elements. It was then possible to calculate the molecular volumes of compounds by adding together the atomic volumes of the elements which they contained. The investigation was continued by Ramsay, Thorpe, Lossen, Schiff, and others; and it transpired that, as a rule, elements in compounds retain the atomic volumes which they possess in the free state; that the constitution of a compound has an appreciable influence on its molecular volume; and that certain elements are capable of assuming different volumes, according to the manner in which they are combined.

Properties which, like molecular weights and molecular volumes, can be numerically valued as the sum of the values of similar terms for elements, are on Ostwald's suggestion

termed "additive." Those which are influenced by constitution, as to some extent molecular volumes, are termed "constitutive;" and those which, like gaseous pressures, depend wholly on the number, and not on the nature of the molecules, are termed "colligative." It was soon discovered that other properties of matter could be treated so as to exhibit their "additive" nature; among these are: the molecular refractive index, or the relative retardation which different compounds and elements offer to the passage of light (Gladstone and Dale, 1858; Landolt, 1864; L. Lorenz and H. Lorentz, 1880; Brühl, 1880); the dispersion of light, or the relative angle which passage through a prism separates rays of known wavelength (Gladstone and Dale, Kanonnikoff); the molecular rotation of the plane of polarized light by its passage through transparent chemical compounds in a magnetic field (Perkin); and molecular coefficients of viscosity (Thorpe and Rodger).

It is, however, also possible to treat such data partly from a "colligative," partly from a "constitutive" point of view; and attempts in this direction, more or less successful, have been made by Schröder and by I. Traube. The latter has been able to apply his method also to molecular volumes in solution in various solvents.

The surface energy of liquids has been shown by Eötvös (1886), and by Ramsay and Shields (1890), to have a simple relation to their molecular weights. At and above the critical point of a liquid it no longer possesses a surface, for liquid and gas are one. As the temperature falls below the critical point, however, the surface energy increases; and the rate of increase for equal numbers of molecules on a liquid surface, is approximately equal for all normal liquids. This affords a means of determining the molecular complexity of compounds and elements in the liquid state, for it is a colligative property. Other methods have yielded evidence corroborative of the quantitative results deduced from measurements of surface tension;

for example, the ratio between the density of a substance at its critical point and its normal gaseous density (Young), the rate of variation of its vapour pressure with increase of temperature, and similar properties.

The distinguishing characteristic of chemical compounds is their constant composition. But it is often difficult to decide whether or not a particular substance has or has not definite composition. Substances which do not exhibit constancy in this respect are termed "mixtures," and such mixtures sometimes consist of compounds mixed with excess of one or other constituent. The investigation of the nature of mixtures, accordingly, has long been regarded as the legitimate sphere of the chemist.

The first definite law regarding mixtures was discovered by Dr. Henry in 1803; it refers to solution of sparingly soluble gases in water. It is: *The amount of gas absorbed or dissolved by a given amount of liquid is proportional to the pressure of the gas.* As, however, the volume of the gas is inversely proportional to the pressure, it follows that a given quantity of liquid always dissolves the same volume of gas, whatever the pressure, temperature, of course, being maintained constant. A convenient way of stating the same fact is to use the word "concentration" to signify the quantity of substance in unit volume; it then follows that the concentration of the gas in the space occupied solely by gas is proportional to the concentration of the solution of the gas in the liquid, for by doubling the pressure the concentration of the gas is doubled, for twice the weight is contained in unit volume; and as the amount in solution is also doubled, its concentration is increased in the same proportion.

Henry's discovery was extended in 1807 by Dalton to the case of gaseous mixtures. His law is: *If a liquid is exposed to a mixture of gases, the amount of each gas dissolved is proportional to its partial pressure, and to its solubility.*

The solubility of gases in liquids generally decreases on

rise of temperature ; but as the critical point of the liquid is approached, other phenomena intervene, which have been investigated largely by Kammerlingh Onnes and his pupils in the physical laboratory at Leiden, in Holland.

The behaviour of mixtures of liquids on distillation has also been investigated by Konovalow (1881); and his work has been greatly extended by Young. In certain cases, if the liquids are immiscible, the vapour pressure at any given temperature is the sum of the vapour pressures of the constituents of the mixture ; if the liquids are miscible, the vapour pressure of the mixture is less than the sum of the pressure of each ; and it may be less than that of either. In certain cases the boiling point of such a mixture remains constant, under constant pressure ; and as constancy of boiling point has been regarded as a sign of definite chemical composition, it has been argued that the mixture was a true chemical compound. But this view is incorrect. Alteration of pressure, as was shown by Roscoe and Dittmar in 1860, alters the composition of the mixture, and the conclusion as to constancy of chemical composition is disproved.

Many researches have been made on the solubility of solids in liquids. As a rule, the higher the temperature the more soluble the solid is. But for each temperature there exists a maximum of solubility ; the solution is then said to be "saturated." Certain substances, however, such as slaked lime, decrease in solubility with rise of temperature ; and this has been shown to be due to the fact that as the temperature rises the soluble compound (in the case mentioned above, calcium hydroxide, Ca(OH)_2) loses water, and is converted gradually into an insoluble, or a much less soluble substance (CaO). The laws relating to the mutual solubility of two or more salts in each other's solutions will be alluded to later.

The state of saturation may be reached in one of two ways : the solution may either be saturated at a high temperature,

and allowed to cool to the required temperature, or the solvent may be shaken up with the powdered solid until it is saturated. In each case the same end-point results; a state of *equilibrium* is reached. Yet Ostwald has shown that if the solid is finely divided, it is more soluble than if in coarse lumps.

The rate of diffusion of liquids was carefully investigated by Thomas Graham (1851). It has already been mentioned that Graham's results as regards the relative rates of diffusion of gases are in accord with the rates of motion of their molecules. The diffusion of liquids into each other is also due to molecular motion. And as gaseous pressure is attributed to the collision of the molecules of the gas with the walls of the containing vessel, so the molecules of liquid, by virtue of their motion, must also generate pressure. But in a mixture of liquids a method for the measurement of the pressure of one of them is not easy to devise. The account of an experiment carried out by Ramsay for gases will render the method clear.

A vessel constructed of the metal palladium, at a high temperature, allows hydrogen to pass through its walls, but is impervious to nitrogen and other gases. If such a vessel be filled with nitrogen, and heated to a constant high temperature under a definite pressure, say that of the atmosphere; and if a current of hydrogen be passed over its exterior, the hydrogen will enter, while the nitrogen cannot escape through the metal walls. The pressure in the interior will rise, owing to the entry of the hydrogen, until the total pressure in the interior of the vessel will equal the atmospheric pressure of the original nitrogen, plus the pressure of the hydrogen which has entered; and if the hydrogen on the exterior is at atmospheric pressure, the total pressure registered will be two atmospheres. The increase of pressure may be regarded as due to the nitrogen, since the hydrogen, being able to pass in each direction through the palladium, will balance its external and internal pressures.

The palladium may be termed a "semi-permeable" septum, since it is permeable to one substance, and not the other.

By a similar device the pressure of substances dissolved in liquids may be measured. In this case the pressure of the dissolved substance is termed its "osmotic" pressure; it is that which tends to spread the dissolved substance through the solution by diffusion, from a place of greater to a place of less concentration; it is due to the motion of the molecules of the dissolved substance.

It had been long known that a bladder filled with alcohol and placed in water would swell, and might even burst, owing to the entry of the water through the animal membrane. This entry was ascribed by F. Perrot (1815) to the tendency of miscible liquids to "wander" through each other, until they were uniformly distributed throughout the mixture. This phenomenon, however, had more direct interest for botanists than for physicists, inasmuch as it appeared to be related to the ascent of sap in plant-stems; and it was first qualitatively investigated by M. Traube (1867), who, after attempting the preparation of artificial diaphragms of tannate of gelatine, and of ferrocyanide of copper, found that these membranes were permeable to some substances and not to others. Quantitative measurements were made by W. Pfeffer (1877), who obtained his best results with ferrocyanide of copper diaphragms; and van't Hoff showed, in 1885, that the quotient obtained by dividing the pressure by the concentration is constant at equal temperatures—a law comparable with Boyle's law for the compressibility of gases, for concentration may be regarded as the reciprocal of volume. Van't Hoff also showed that the increase of osmotic pressure with increase of temperature followed Gay-Lussac's law, and again confirmed the analogy between gases and substances in dilute solution. De Vries and Tammann amplified the work of Pfeffer (1888) by determining at what degree of dilution various solutions exercised

equal osmotic pressure; and van't Hoff pointed out that for certain substances, such as sugar, salicin, etc., the quantities of dissolved substances were then present in proportion to their molecular weights. Moreover, he went a step further and showed that the osmotic pressure exercised by a solution of cane-sugar at a definite temperature and at a definite concentration (*i.e.* with a definite weight of cane-sugar contained in unit volume of the solution) was the same as the pressure which would be exercised at the same temperature by a gas containing a number of molecules per unit volume equal to that of the cane-sugar.

The osmotic pressures of dissolved salts, however, do not exhibit the same simplicity. The reason for this divergence was first pointed out by van't Hoff, and will be referred to later.

The fact that solutions of compounds in pure solvents raise the boiling point of the solvent had been known for long; it was investigated in 1822 by Faraday, and later (1824) by Griffiths, and (1835) by Legrand. They, and Wüllner (1856-1858), investigated chiefly the behaviour of salts; and Ostwald (1884) pointed out that if the rise in boiling point of the solution be referred, not, as these authors had done, to the relative *weights* of dissolved substance, but to their relative *molecular weights*, equal numbers of molecules of different salts, dissolved in the same weight of solvent, produce an approximately equal rise in the temperature at which the solution boils. But it was not until Raoult (1886) had extended the researches to solutions of carbon compounds other than salts, that regularity was introduced. It then transpired that equal numbers of dissolved molecules produce an equal rise in the boiling point of the solution; or, stated in another manner, the presence of equal numbers of molecules of two substances in equal quantities of the same solvent reduces the vapour pressure of the solutions to an equal extent. The connection between these

phenomena and between similar relations dealing with the depression in the freezing point of a solvent produced by the pressure of dissolved substances was shown by van't Hoff in 1887; and the thermodynamic explanation was given by him, and in a different form also by Arrhenius in 1888.

Arrhenius, too, propounded a theory to account for the irregular behaviour of salts as regards their osmotic pressure, and the rise in boiling point and depression in freezing point of solutions containing them, which they exhibit. It is—that when dissolved, salts dissociate into two or more constituents, each of which produces an effect as if it constituted a separate molecule; and that these constituents are identical with the *ions*, or carriers of electricity, when the salts are subjected to the influence of an electric current. Many arguments in favour of this hypothesis were collected and published by van't Hoff in 1887; others are to be found in the volume-relations, the colour, the optical, the magnetic, and other properties of salts.

The connection between the osmotic pressure of dissolved substances and the rise of boiling point or depression of freezing point which their solutions exhibit can be shown only by the aid of thermodynamics. But one consideration will make it plain that such a connection must exist. The osmotic pressure of a dissolved substance evidently depends on its concentration in the solution; if the concentration be increased, the osmotic pressure will be increased proportionally. This could be achieved by applying an external pressure to the solution, so that solvent would be expelled through the semi-permeable membrane, just as the pressure of a gas can be increased by diminishing its volume by application of external pressure. Now, the amount of work done in diminishing the volume of the solution is the same, whether the solvent be removed by pressing it through the diaphragm, or by freezing it out, or by removing it by evaporation; and thus the relation between

osmotic pressure and the depression of vapour pressure or freezing point can be established. There is, indeed, a fourth method of altering concentration, and that is by shaking the solution with some other liquid which does not mix with the original solvent, but which dissolves the substance, and so removes it partially from the original solvent; and here, again, similar thermodynamic considerations can be introduced.

The relationship between the volumes, expansions, optical properties, and other properties of solids have been to some extent worked out. Many approximate numerical relations, based on the molecular weights of the solids, have been elucidated; but, as a rule, the results have not been so useful as those with gases and liquids. Allusion has already been made to the equality of the atomic heats of the solid elements, discovered by Dulong and Petit; and to the extension of Dulong and Petit's law to compounds by Neumann in 1831. Regnault, Kopp, and Joule have contributed to our knowledge of such relations; the ease with which very low temperatures can now be obtained leads to the hope that in the near future this knowledge may be considerably increased, and that some philosophical basis may be found for the irregularities which have hitherto been discovered.

That almost every change in a chemical system is accompanied by a gain or loss of energy is evident, if heat be regarded as a form of energy, for heat is almost always evolved or absorbed during the formation or decomposition of every chemical compound. The total energy in any substance eludes measurement; but the difference in energy-content between any one state and any other can be determined. The province of thermal chemistry is the measurement of such differences of energy in the form of heat.

The first important theoretical advance in this direction was due to Hess (1840), in his postulate that the total *evolution*

of heat during any chemical process is the same whether the process takes place in one or in several stages. Actual determinations of the heat evolved during combustion of various substances, and of the heat-changes which take place on solution, were made by Andrews (1841 and subsequent years) and by Graham (1843); and Favre and Silbermann greatly added to our knowledge of facts of this nature (1852-1853). The most remarkable series of researches on the heat-changes which accompany chemical action are due to Julius Thomsen of Copenhagen, and to Marcellin Berthelot of Paris; the former published his first treatise in 1853, and the latter in 1865, and they have continued their investigations almost to the present day (1903).

Each of these authors enounced a principle, which has subsequently turned out to be incorrect, viz. that those chemical reactions take place which are accompanied by an evolution of heat. But this incorrect hypothesis led to the production of an enormous mass of experimental work. The evolution of heat during the formation of numerous compounds from their elements, the heat-changes occurring during the conversion of one compound into another, the heats of combustion of compounds of carbon and other elements, that evolved during the neutralization of bases by acids and during the progress of allotropic and isomeric change, have, in many cases, been measured, and many interesting conclusions have been arrived at. One of the most interesting is the fact that on neutralizing a dilute solution of a monobasic acid by a monoacid base, nearly the same amount of heat is evolved in most cases; and the tracing of this fact to its cause—that such an action is accompanied by only one chemical change, the combination of the hydroxyl ion, OH' , of the base with the hydrogen ion, H' , of the acid. The metallic and acid ions remain, for the most part, unaffected by mixing their solutions. When combination occurs, or where the acid or the base, in solution, exists not wholly in the state of ions, the regularity no longer holds.

Other interesting results have been arrived at in the case of explosive bodies, which are always endothermic, *i.e.* are formed with absorption of heat; while stable bodies are generally produced with heat evolution. With rise of temperature, substances formed with evolution of heat become more unstable; and the converse is also true, that endothermic substances become more stable on rise of temperature.

On the whole, however, little of theoretical importance has been deduced from the very numerous data of thermal chemistry. It is true that F. W. Clarke has recently published the outlines of an attractive theory, which makes it possible to calculate the heats of formation of many compounds from simple considerations; but the accuracy of Clarke's deduction has been disputed by Julius Thomsen.

The first application of electricity to chemistry dates from the time of Priestley (1772); he proved that the effect of passing electric sparks through air was to produce an acid substance, which he supposed to be carbonic acid; but in 1775 Cavendish carried out a marvellously careful set of experiments, and showed that the acids actually produced were nitrous and nitric. Not long afterwards van Marum (1785) and van Trostwyk and Deimann (1789) showed that hydrogen and oxygen result from the action of powerful sparks on water.

Shortly after (1791) Galvani discovered that a feeble current of electricity caused the muscles of a frog's leg to contract, and Volta invented his "pile." With such a "pile," Nicholson and Carlisle "decomposed water" (1800), and found that from the wire attached to the zinc disc oxygen was evolved, while from the other wire bubbles of hydrogen escaped. At the same time the water round the former became acid, and round the latter alkaline. That acid and alkali could be produced by the decomposition of "pure" water was a puzzle to many; and it was not until Humphry Davy showed in 1800 that

the alkalinity and acidity were due to the electrolysis of minute traces of salt, dissolved out of the vessel in which the water was contained, and that pure water in vessels of gold or platinum was a practical non-conductor, and gave rise by its decomposition to no alkali or acid, that the anomaly was explained.

Davy also evolved a theory by means of which he endeavoured to connect electrical with chemical phenomena. Starting from the known fact that acids or bases in contact with metals acquire an electrical charge, he argued that the same must be the case with atoms ; that in entering into chemical combination they acquire electrical charges of different sign, and cohere, owing to the same reason which causes electrical attraction of oppositely electrified bodies. According as the charge is greater or less, the chemical attraction is powerful or feeble. Davy found the difference of chemical potential to increase with rise of temperature, and drew the not unnatural conclusion that this increase runs parallel to the increase of chemical affinity, which causes combination more readily at a high than at a low temperature. Indeed, Davy did not regard these as two different phenomena, one the cause, the other the effect, but he regarded them as essentially the same.

Decomposition by an electric current he believed to be a reversal of the phenomena of combination. In giving the atoms of the decomposing compound opposite charges to those which they had acquired by their combination, the atoms were restored to their previously neutral state ; the positive atoms or groups were attracted to the negative pole ; their positive charges were neutralized by a corresponding negative charge from the pole, and the atom or group was liberated in the free state.

Davy's views made no lasting impression, for they were shortly afterwards superseded by those of Jacob Berzelius (1806). Berzelius held the view that all chemical compounds consisted of a combination of two opposite constituents. To this view he was led by the observation that an electric current

apparently decomposed salts into an acid portion and a basic portion, each of which goes respectively to the negative or to the positive pole ; combustible substances, alkalies or alkaline earths, collect at the positive pole, while oxygen, acids, and in general oxidized substances go to the negative pole. He also believed that the decomposition of mixtures or of compounds was in compound proportion to their chemical affinity and to their surface of contact, and that the absolute amount of the decomposition was proportional to the quantity of electricity, and that this varied with the surface of contact between the metal and the conducting liquid ; it was also supposed by him to be proportional to the conductivity of the solution. The course of the reactions occurring during decomposition was believed to be influenced by the affinity of the components for the material of the wires, by the mutual affinity of the constituents, and by the cohesion of the resulting compounds. To justify these suppositions he made the hypothesis that each atom contains electric charges of different amounts ; and in general both a positive and a negative electric charge, one at each pole. His theory differed from that of Davy essentially in this, that while Davy believed that the atoms acquired their charges only by contact with each other, Berzelius imagined the atoms to be endowed from the start with electrical charges. But although any atom contained both a positive and negative charge, they were not necessarily equal in amount ; hence some atoms were essentially negative, whilst others were essentially positive. During combination, the oppositely electrified atoms attracted each other, and by equalizing their charges, more or less, heat and light were the accompaniments of the partial or complete restoration of the electrical neutrality. Berzelius himself acknowledged, however, that the neutralization of the electric charges removed the cause of the persistence of the compound, if chemical affinity were considered to be due to electric attraction. But Berzelius made use of his "binary"

theory more as a means of classification than for the purpose of explaining chemical or electrical phenomena ; and owing to that advantage it persisted for long.

The next advance in the subject was due to Michael Faraday (1831-1838). His first statement was : *The amount of a substance decomposed by an electric current is proportional to the amount of electricity which passes through it.* This was soon after followed by the important law which goes by his name : *If the same current passes through several electrolytes, the amounts of the different substances separated from the compound are proportional to the chemical equivalents of the substances separated.* The same amount of electricity, he showed, always liberated the same amount of explosive mixture of oxygen and hydrogen from dilute sulphuric or hydrochloric acid, whatever the "strength" of the current, the size of the electrodes, the dilution of the liquid, or the temperature.

It was Faraday who introduced the term *electrolysis* to signify the process of separation of the constituents of a compound body by means of an electric current ; the conductor, of which the components undergo such separation, he called the *electrolyte* ; the moving parts were termed by him *ions* (or "movers"), and those which go towards the *anode* he called the *anions*, and similarly, the ions which move towards the *kathode* were termed by him the *kations* (*ana* = up ; *kata* = down ; *hodos* = a way).

Faraday was careful to point out that the passage of a current through an electrolyte, and the separation of the ions at the electrodes, although they are concurrent, are yet absolutely distinct phenomena.

While Davy's view that the products of electrolysis are the metal or hydrogen, on the one hand, and the non-metal, or electro-negative group, on the other, had been overshadowed by Berzelius's contention, that one product is an acid, and the other a base, the question was not settled until Daniell,

Professor at King's College, London (1839), showed that if a current be passed through "water" (dilute acid) as well as through a solution of sodium sulphate, the same amount of hydrogen is collected in each case; but in the latter, an equivalent of "soda" (sodium hydroxide) is also set free. Hence the current would appear to be doing double work in the solution of sodium sulphate; not merely liberating the correct amount of hydrogen, but also liberating the soda. He pointed out the obvious explanation: that the primary product of the electrolysis of the sodium sulphate is sodium, and that the hydrogen is really produced by the action of that metal on the water, at the moment of its liberation.

Early in the days of electro-chemistry (1805) a hypothesis was brought forward by Grothuss to explain a phenomenon which had been pointed out by Davy, namely, that the element liberated at one pole has not been driven bodily from the opposite pole to the one at which it is liberated. As a striking proof of this, he caused the current by which "water" was electrolysed to pass through his own body; and it was inconceivable that the same atom of oxygen, for example, could have passed from the negative to the positive pole through his organism. Grothuss imagined the atoms to be already charged. The atoms of oxygen and hydrogen in the water molecules gave up their charges to their respective poles, and escaped; the neighbouring atoms changed partners, those at the end of the chain combining with the atoms left without partners. But Clausius (1857) disproved Grothuss's theory, which had maintained its ground for over half a century. He pointed out that if a certain "force" is required to effect decomposition, no decomposition should take place until that force has come into operation; as a matter of fact, however, the smallest conceivable electric "force" is sufficient to effect decomposition of an electrolyte. He therefore imagined that in any electrolytic solution some of the atoms are continually changing partners,

and that they move under the influence of the current at the instant when they are unattached, that is, while the exchange is taking place. This hypothesis had previously been put forward by Williamson, with the view of explaining all chemical action; some molecules, he supposed, were always undergoing decomposition; their constituent atoms, or groups, however, did not necessarily reunite, but were ready to unite with those of other molecules which also had undergone decomposition.

The relative rate of motion of the ions was first measured by Hittorf. Although it had been noticed by Faraday (1834), and by Daniell and Miller (1845), that during the electrolysis of a salt the salt became more concentrated in the neighbourhood of one pole, and more dilute round the other, it was not until 1853 that the true explanation was found by Hittorf in the fact that the two ions do not move at an equal rate. If they do, there is no such alteration of concentration. But in the more numerous cases, where their rate of motion is different, the salt concentrates round the pole to which the more rapidly moving ion is travelling.

Hittorf also pointed out that compounds, solutions of which are good conductors of electricity, are precisely those bodies which are most susceptible of chemical reaction. In later years this dictum has proved one of great importance.

Accurate measurements of the rate of passage of charged ions through their solutions were first made by Kohlrausch in 1869, with this remarkable result: that the ions move independently of one another. Hence the electrical conductivity of any salt may be arrived at by the *sum* of the rates of transport of its ions.

Further progress was made in 1887 by Arrhenius, who showed that, on sufficient dilution, all salts arrive at a maximum conductivity, of the same order of magnitude; and he deduced from this that, with increasing dilution, a larger and larger number of molecules become ionised, *i.e.* resolved into ions.

Indeed, many salts, in fairly concentrated solution, are already largely split into ions; while acids, on the other hand, even at very small concentration, are to some extent in the non-ionised form. The reactivity of a salt, he contended, was due to the actual number of ions present in unit volume. And van't Hoff showed the proportionality between the relative number of ions, thus determined electrically, and the osmotic pressure which they exert, as well as their influence in raising the boiling point and depressing the vapour pressure or the freezing point of the solutions which contain them.

The connection between the amount of energy expended electrically in the cell of a battery, and the heat evolved when the reaction between the constituents of the cell proceeds without production of a current, or with a closed circuit, was first calculated and experimentally proved by Joule and Thomson (Lord Kelvin), the latter of whom summed up the results of the investigation in the proposition: *The intensity of an electro-chemical apparatus is, in absolute measure, equal to the mechanical equivalent of as much of the chemical action as occurs by means of a current of unit strength, in unit time.* But experiments by Boscha, Raoult, and others did not always succeed in corroborating the truth of this assertion. The anomaly was explained by Helmholtz, as well as by Willard Gibbs (1878) in his great work. The discrepancy was caused by the neglect to take into account the temperature-coefficient of the electromotive power of the particular cell employed. If this is small, as, for example, in a Daniell's cell, in which zinc is replaced by copper, the agreement with Joule and Thomson's statement is a close one; but if it is large, as in many other instances, the necessary correction must be introduced in order that experiment and theory give the same result. The further development of electro-chemistry belongs to the present day, and will be thoroughly considered in the volume devoted to that subject.

The connection of chemical action with light is a subject which has attracted much attention, owing to its connection with the practical aspect of photography on the one hand, and with astronomy on the other, in the form of spectroscopy. But even yet few general conclusions have been drawn.

The fact that silver-salts are alterable by light was observed as early as 1727 by Schultze; and Scheele, the discoverer of oxygen, observed that this action is due chiefly to the violet part of the spectrum (1777). Senebier made quantitative experiments in 1784; and Daubeny, in 1836, made similar quantitative experiments on the influence of different colours of the spectrum on the absorption of carbon by plants.

It was for long believed that three distinct varieties of waves were emitted by luminous bodies; waves of light, of heat, and of chemical action; but it appears that all waves may have all three effects, and that the wave-length of those waves which produce the maximum of chemical action depends on the kind of chemical action which results from them. Thus, while the maximum effect on plant-life is produced by light of such a wave-length that it has also nearly the greatest luminosity, the maximum effect in decomposing silver bromide is caused by vibrations in the ultra-violet part of the spectrum; and heat effects are produced by vibrations in the infra-red.

It has been pointed out by Mayer that while, generally speaking, the effect of ether waves in promoting chemical action is merely that of causing it to start, so that energy is lost, the action on living plants is of an opposite character, for by its influence energy is stored. The action in the first case is one resembling catalysis, and merely increases the rate at which chemical action proceeds; while in the second, the radiant energy from the sun is stored by means of the plant as chemical energy.

Many attempts, more or less successful, have been made to measure, by the chemical changes produced, the intensity of

the radiations which produce these changes; the best known are the researches of Bunsen and Roscoe (1851-1862), and Draper (1857). Many more recent researches have, however, been made.

The development of spectroscopy has of recent years been very rapid, so far as relates to the measurement of the spectra of different substances. Although the refraction of light was first explained by Newton, and investigated in the early half of the eighteenth century by Brewster, rapid progress was made when Kirchhoff discovered, in 1860, that all substances emit rays of light of the same wave-length as those which they absorb. The introduction of the spectroscope as a means of analysis by Bunsen and Kirchhoff drew attention to the advantage of the study of spectroscopy, and established many important laws. The fact that gases made luminous by an electric current of high potential could be made to emit different kinds of spectra, according to the nature of the discharge, was the discovery of Plücker and Hittorf (1865); and the fact that each compound, if it can only stand the temperature at which it emits luminous rays, has its own special spectrum was first announced by Mitscherlich in 1869.

The first theoretical discussion of spectra is due to Maxwell (1875); he pointed out that harmonic relations should be found between the wave-length of the lines of the spectra of any substance. Mascart, Stoney (1871), and Lecoq de Boisbaudran (1889) have shown that such "harmonic relations" exist in some cases; and other authors have sought to establish such relations; the most successful is due to Balmer, whose formula has yielded excellent results in the hands of Rydberg, Runge and Paschen, Kayser, Lorenz, Zeeman, and others.

There is, however, a vast unexplored field in spectroscopy; for although the visible waves, as well as those in the ultra-violet part of the spectrum, have been investigated in many instances, the infra-red waves have been hardly looked for.

Langley, indeed, has measured the lines in the infra-red part of the solar-spectrum by means of his "bolometer," an instrument capable of detecting very minute differences of temperature; but this instrument has been little applied to the spectra emitted by gases or by hot solids.

Speculations regarding the reason of chemical combination date from the earliest times. The word "affinity," or "chemical affinity," calls us back to the time of Hippocrates, whose view was that "like draws to like;" that substances which combine must have something in common, like wine and water, silver and gold. As solution and mixture passed in those days for combination, there was some reason for his dictum. Empedocles, the first atomist, held a different view, founded on imaginary hates and loves between the atoms. In the Middle Ages, even quainter ideas are to be met with; that the particles of the acid solvents are sharp, like needles or spears, and that they transfix and support the particles of dissolved metal; that sweet liquids are sweet, because of the rounded form of their ultimate particles; and that they therefore have not a similar solvent action. With Newton a new view arose. He himself did not believe that affinity was due to an attraction identical with that of gravitation; but leant to the view that the force varied inversely with some higher power of the distance than the square. But Buffon, the French naturalist, in spite of Newton's well-considered thought, supposed that the form of the objects attracting—the ultimate particles—could make it possible that the laws of the inverse square held for chemical attraction; and his views were widely disseminated and accepted.

Davy's and Berzelius's electro-chemical theories have already been alluded to, and need not be further discussed here.

Attempts to arrange qualitatively the order of affinity of the elements for each other were frequently made in the eighteenth

century. As early as 1718, Geoffroy arranged in order various chemical substances (for elements in the modern sense of the word were even then not recognized) in the order of what he supposed to be their affinity for each other. For instance, under "metallic substances" we find the order—iron, copper, lead, mercury, silver, as regards their affinity for nitric acid; and for vitriolic acid the order was—sulphurous or oily principle, fixed alkali, volatile alkali, absorbent earth (*i.e.* calcium carbonate), iron, copper, silver, and so on. The idea which guided him he expressed thus: "Whenever two substances, which have some inclination to combine, are combined with each other, and a third is presented to them which has more affinity to one of the two than the other, that other is expelled." The same idea was fully elaborated by Torbjorn Bergmann in 1783. He distinguished between various kinds of attraction: for example, particles of the same kind were held together by *attractio aggregationis*; compounds remained compounds by virtue of *attractio compositionis*, which might be of two kinds, according as they had been prepared in solution (*attractio solutionis*) or by fusion (*attractio fusionis*). Simple expulsion of one constituent out of a compound by the action of another was due to *attractio simplex electiva*; while exchange between the constituents of two compounds was caused by *attractio duplex*. Attempts not merely to determine the order, but also the relative amount of such affinities, were made by De Morveau (1793); he attempted to ascertain the relative affinities of metals for mercury by measuring the weights required to separate plates of a number of metals of the same size from a surface of mercury, and concluded that as adhesion and affinity must be fundamentally the same, he was measuring relative affinity. He also gave six laws of affinity: that only liquids act; that affinity takes place only between the smallest particles of substance; that the affinity between any two substances is partly conditioned by the relative amount of each; that chemical affinity acts only

if it can overcome cohesion ; that compounds differ entirely in properties from their constituents ; and that affinity is greatly influenced by temperature.

We have here a statement that the relative amounts of substance present influence their behaviour as regards a third. This law was originally stated by Wenzel in 1777, in this form : The affinities of bodies for a common solvent are inversely as their rates of solution in that solvent ; also, that the strength of chemical action is proportional to the concentration of the acting material. Count Berthollet, independently of Wenzel, enunciated the same doctrine in 1799, and stated clearly that it was impossible, as Bergmann had attempted, to arrange substances in a definite order of chemical affinity, but that the affinity was deeply affected by the relative amounts of substances present. He succeeded in reversing the actions which had hitherto been supposed to decide the order of chemical affinity : for example, he decomposed barium sulphate by boiling potash ; potassium sulphate by boiling milk of lime ; calcium oxalate by nitric acid, and so on. Although in each case only a partial decomposition was attained, yet that decomposition occurred at all was regarded by him as a proof that a large quantity of a reagent could reverse the order of chemical affinity. He regarded these reversed actions to be due to the small surface of the precipitate, compared with the large and constantly renewed surface of the acting liquid ; so that when a liquid acts on a solid, the activity of the liquid is conditioned not by its total amount, but by the degree of its concentration ; and similarly for gases. For when a gas is liberated by any reaction, it removes itself, and can no longer act on the dissolved substances ; hence a comparatively small quantity of reagent is capable of liberating a large quantity of gas. The elasticity of the gas may be regarded as a force acting in opposition to the affinities of liquids. Combination may be either helped or hindered by a rise of temperature ; helped, if

the rise of temperature so diminishes the cohesion of the acting substances as to increase chemical action; hindered, if one of the substances changes its state by the rise of temperature and removes itself from the sphere of action. The action of solvents is in reality the overcoming of cohesion, so that the affinities of the dissolved substances can manifest themselves.

It is thus evident that Berthollet's ideas on the measurement of chemical affinity differed in important respects from those of his predecessors. According to him, at least one of the reacting substances must separate, in order that a reaction shall take place; the state of equilibrium is thus destroyed, and the reaction proceeds until so much of one or other substance separates as to restore the balance.

But Berthollet was misled into making the false statement that the composition of compounds may vary within limits, and that the proportion of the constituents of each compound depends on the relative proportion in which the constituents are present during the process of its formation. Proust, a countryman of Berthollet's, refuted Berthollet's statement by direct experiment, and showed that the supposed compounds on which Berthollet based his contention were not really definite compounds, but mixtures. The definite and invariable composition of chemical compounds, as has been already stated, was fixed by Dalton and by Berzelius.

While, then, it is true that the relative amount of a compound formed during any reaction is conditioned by the proportions of the reacting substances present, it does not follow that the composition of the compound itself is thus affected; nor is it true, as Berthollet fancied, that the relative affinities of acids are to be gauged by the relative amounts required to saturate a given base. While Gay-Lussac saw this error, he defended Berthollet's main contention, and maintained that the partition of acids with a base sufficient to saturate only one of them was affected by the relative amounts of the acids

present. But the difference between altering the relative amount of a compound formed and altering the composition of the compound is a fundamental one.

Berthollet's work, *Essai de Statique Chimique*, was consequently soon forgotten; and it was not until 1835 that the question was revived by Persoz, Professor at Strassburg, who attempted to give a table analogous to Bergmann's of the relative affinity of oxides for acids. And Berthollet's views were resuscitated for analytical purposes by Rose in 1842, and later, in 1853, by Bunsen. Bunsen exploded a mixture of hydrogen and carbon monoxide with an amount of oxygen insufficient for both, and concluded that instead of a gradual progression in the proportions of water vapour and carbon dioxide produced, the ratio remained definite until a certain excess of oxygen was present, and that it then altered suddenly; so that there was always a definite molecular proportion between the amounts of the two products of oxidation. Similar results were obtained by his assistant Debus, in precipitating carbonates of calcium and barium by the action of carbon dioxide or their hydroxides. But these experiments were subsequently proved to be erroneous.

The law of "mass-action," although foreshadowed by researches of Biot (1835) was first stated in precise terms by Wilhelmy in 1850. In order to examine the state of equilibrium in any system, it is clear that the conditions must not be changed: for example, the solution must not be concentrated by evaporation; nor must other substances be added; nor must the temperature be altered; nor must the solvent be changed, as, for example, by the addition of alcohol to an aqueous solution; hence some physical property must be made use of, which shall reveal the result of the change which has taken place, without disturbing equilibrium.

For this purpose Wilhelmy made use of the rotation of the plane of polarized light caused by cane-sugar, and by its products

of decomposition on treatment with an acid, dextrose and levulose. In this way the rate of change could be measured without the introduction of any foreign substance. This law and its consequences will be fully considered in the volume on chemical dynamics. In 1855, J. H. Gladstone made use of the colours of solutions of reacting substances and their products to investigate the position of equilibrium, and in one instance he employed the rotation of polarized light; and to him belongs the credit of pointing out that after this manner tables of relative affinity could be constructed. Berthelot and Péan de St. Gilles, in 1862 and 1863, in investigating the action of acids on alcohols, which is attended with the formation of esters, did much to elucidate the nature of chemical action. They showed that the action is a gradual one; that it is never complete, but that the amount of ester formed tends towards a limiting value; and that this limit is the same, whether it be reached by the action of acid on alcohol, or of water on ester. They also showed that the rate of action, but not the limit, was greatly increased by rise of temperature, and that pressure has little influence on the process. Their conclusion agreed with that of Wilhelmy, that the quantity of ester formed at any moment is proportional to the product of the acting masses, and inversely proportional to the volume they occupy.

About this date (1857) Deville began to publish his results of experiments on dissociation, which have already been alluded to. He drew an analogy between the temperature of dissociation of a compound and the boiling point of a liquid; or, to be more precise, the pressure exercised by a gaseous constituent of the dissociating body at a given temperature, and the pressure exerted by the vapour of a liquid at a given temperature. With rise of temperature, vapour pressure increases; and similarly with rise of temperature the pressure due to dissociation increases. Curiously enough, however, Deville was against the theory of mass-action; and yet his

experiments furnish one of the most striking proofs of the justice of that doctrine.

The first attempt to determine the value of chemical affinity by help of the then recently developed ideas of thermodynamics is due to Julius Thomsen, of Copenhagen (1854). Defining "affinity" as that force which holds together the constituents of a compound, he believed that it was possible to measure the amount of that *force* by measuring the *heat* evolved in a reaction by which the body was formed, which would be equivalent to the heat required to decompose the body—that is, he thought it possible to measure a *force* in terms of an *energy*. He also believed that all chemical changes which take place spontaneously, or after being started, are necessarily accompanied by an evolution of heat. It is true that he was acquainted with changes, such as the solution of salts in water, which were attended by absorption, and not evolution of heat; but he attempted to explain away such cases by premising that it was only purely chemical actions which were attended by evolution of heat.

In 1867, Berthelot independently announced the same principle; and a somewhat heated discussion between Thomsen and Berthelot took place in 1873, as to claims of priority and as to the exact nature of the statement involved, which need not be further pursued, inasmuch as the matter in dispute was a false statement.

But the question had been solved by Waage, former Professor of Chemistry, and Guldberg, Professor of Mathematics, at Christiania, who published, in 1865, a pamphlet termed *Études sur les Affinités chimiques*. The fundamental idea of this work was the law of mass-action: that *the chemical action is proportional to the quantities of substances acting; the last being determined by the amounts in unit volume*. If two substances act on each other, the action is proportional to the relative amount of each, and will be zero if the amounts of acting bodies is zero,

and therefore it is proportional to the product of the two. Further, the action will depend on the nature of the substances, on the temperature, and on other circumstances; these influences can be expressed by a factor, hence the "force" of the chemical affinity can be represented by the expression kpq , if k is the factor, and p and q are the acting masses. But this action is balanced by forces acting in opposition to the formation of the new system, which tend to restore the primary condition of the substances. If the substances formed by the original reaction are p' and q' , and the new factor k' , equilibrium will be established when $kpq = k'p'q'$. Although it is not possible to calculate the actual amount of the "chemical forces," it is possible to calculate their ratio, by knowing the position of equilibrium, when the reaction has gone as far as it will, under a given set of conditions. The actual result of such calculations is, however, to give the relative velocities of chemical reactions; and van't Hoff, in 1877, pointed out that it is sufficient to confine attention to such velocities, and not to ascribe the velocities to an impelling force, and thereby to introduce mechanical ideas which are unnecessary for the purpose. Guldberg and Waage proved their thesis by many experimental researches; while in 1869 Julius Thomsen had shown that the problem can be solved by thermochemical measurements. Ostwald, in 1876, made quantitative measurements of volume-changes during reactions, which also afforded data for the proof of the law.

Horstmann was the first to apply thermodynamics to chemical processes (1869), treating first the problems of dissociation. The similarity between dissociation and vapour pressure makes it possible to treat the former in the same manner as the latter, because both classes of reaction are reversible. Berthelot's and Thomsen's law, implying that that change necessarily takes place which is accompanied by a dissipation of *energy*, was replaced by another: *that of all possible*

systems that one is most stable in which the entropy is at its maximum. The consideration of this doctrine will be found in the volume on Thermodynamics. This principle was later announced by Lord Rayleigh. The most remarkable advance in this direction, however, is due to Willard Gibbs (1874-1878). It is hardly too much to say that all subsequent advances are merely applications and variations of Gibbs's fundamental considerations. In his epoch-making work, Gibbs treats of the chemical potential of a substance, of coexisting phases of matter, of cases of mixture of ideal gases as regards their equilibrium with liquids and solvents, or with solids, of surface-tension, and of electric action between bodies.

In van't Hoff's *Études de dynamique chimique* (1884), numerous instances are adduced of the application of the laws of chemical dynamics, and the influence of temperature was especially studied. And after Arrhenius of Stockholm had brought out his theory of the independent existence of ions in solutions (1884), van't Hoff in 1887, as has already been narrated, drew deductions from measurements of osmotic pressure by de Vries and by Pfeffer, of the depression of vapour pressure and of rise of boiling point of solvents caused by dissolved substance, and of lowering of freezing points investigated by Raoult, and of Arrhenius's determinations of electric conductivity, and welded all these ideas into a homogeneous whole.

The present volume, which forms the first of the series of works dealing with physical chemistry which it is proposed to publish, treats of the Phase Rule, one of the subjects treated of by Willard Gibbs, in which many of the considerations touched on in the foregoing pages are introduced.

It is proposed, in subsequent volumes, to treat of *Stoichiometry*, or the various methods employed to determine atomic and molecular weights, and the classification of compounds (Professor Sydney Young, D.Sc., F.R.S.); of the Relations

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